Pd/Cu site interchange in $UCu_{5-x}Pd_x$

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Abstract

Although Pd/Cu site interchange in the non-Fermi liquid (NFL) material UCu_4Pd has been observed, the relationship between this disorder and the NFL behavior remains unclear. In order to better compare to the $UCu_{5-x}Pd_x$ phase diagram, we report results from Pd K edge x-ray absorption fine structure (XAFS) experiments on this series ($x = 0.3 - 1.5$) that determine the fraction of Pd atoms on the nominally Cu ($16e$) sites, s. We find that for these unannealed samples, s is at least 17% for all the samples measured, even for $x < 1.0$, although it does climb monotonically beyond its minimum at $x = 0.7$. These data are compared to changes in the lattice parameter as a function of x.

Key words:
non-Fermi liquid, lattice disorder, XAFS

UCu_4Pd was thought until recently to be an example of a well-ordered NFL material, but actually has a significant ($\approx 1/4$) fraction, s, of Pd's on nominally Cu ($16e$) sites [1]. The existence of this site interchange in UCu_4Pd originally prompted us to conjecture that some bond length disorder should also exist [1], and we showed that it is then possible to model the magnetic susceptibility with the ensuing distribution of Kondo interactions. Evidence of a spin glass phase has been observed near 200 mK in some UCu_4Pd samples [2,3], lending further credence both to the existence of lattice disorder, the possible bond length disorder, and to the notion that this disorder affects the magnetic properties. However, recent annealing studies of UCu_4Pd [4] have shown that the lattice parameter decreases when the samples are annealed

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appropriately, and that the logarithmic divergence in the heat capacity (a NFL hallmark) then extends over a wider range. The decrease in the lattice parameter with annealing is thought to be due to a decrease in the Pd/Cu site interchange. These measurements therefore argue against a Kondo disorder-type mechanism [5] for the NFL behavior. Together with measurements of scaling in the magnetic response [6], the situation remains muddled. Does the lattice disorder serve only to create a mixed-phase system, where the spin glass behavior is associated only with the disordered phase and the NFL behavior is due to a well-ordered phase that exists near a quantum critical point [7], or does the disorder precipitate a broader range in phase space for quantum critical behavior to exist, as in a Griffiths' phase, either through the competition between Kondo and RKKY interactions [8] or from Anderson localization [9]?

To explore some of these issues and to try to better understand the relationship between the lattice parameter and the site interchange s, we have measured s as a function of x in $UCu_{5-x}Pd_x$ up to x=1.5 as part of a larger study [10]. The samples were prepared by arc-melting appropriate amounts of the end members UCu_5 and UPd_5 in an ultra-high purity Ar atmosphere. The samples were not annealed. X-ray powder diffraction measurements reveal the compounds crystallize in the $AuBe_5$ structure with similar lattice parameters to those of Refs. [3,4] and no traces of impurity phases were observed within the resolution limit of the experiment. Site interchange is measured using the x-ray absorption fine-structure (XAFS) technique from the Pd K edge, as described previously [1]. A more complete model for determining s is now employed, and will be describe elsewhere [10].

Figure 1 shows the Fourier transforms of the XAFS function $k^3\chi(k)$ from some of the XAFS measurements, which roughly correspond to the radial bond length distribution around the average Pd site for each sample. Please note that the peak positions are shifted from the actual bond lengths due to a phase shift of the photoelectron in the vicinity of the absorbing and backscattering atoms. In any case, the main utility of XAFS for measuring site interchange in these systems comes from the difference in the near neighbor environment around the nominal Pd ($4c$) and Cu ($16e$) sites. In the ordered x=1 system, the nearest neighbors to Pd are 12 Cu atoms at ≈2.93 Å. If any Pd atoms rest on $16e$ sites, the nearest neighbors to these Pd's are 6 Cu atoms at ≈2.55 Å. Therefore, Pd/Cu site interchange is manifest primarily as a shoulder on the low-r side of the peak in Fig. 1 corresponding to the 2.93 Å Pd-Cu pairs, which are at ≈ 2.6 Å in the Fourier transform. Detailed fits [10] give the actual measurements of s, as displayed in Fig. 2a.

The first point to note is that $s > 0$ for all measured x's, although the error bars on the x=0.3 measurement may be consistent with $s = 0$. In addition, since UCu_5 is known to oxidize readily,[11] it is possible that the poorer quality of the fit is caused by a small amount of UO_2. The minimum measured s is

at $x = 0.7$. At larger x's, s begins to rise monotonically. In a rough sense, s approximately follows the nominal concentration, except the rise in site interchange begins at $x = 0.7$ instead of $x = 1.0$ and there is an offset of approximately 0.2 in s. This region of $x = 0.7$ is special for other reasons. For one, samples with $0.7 < x \lesssim 2$ display NFL behavior. Also, for *unannealed* samples the lattice parameter a begins to increase more rapidly with x above 0.7 than below. Therefore, the increase in a is likely due to the increase in s. This idea is consistent with the interpretation by Weber *et al.* in their annealing study of UCu_4Pd [4].

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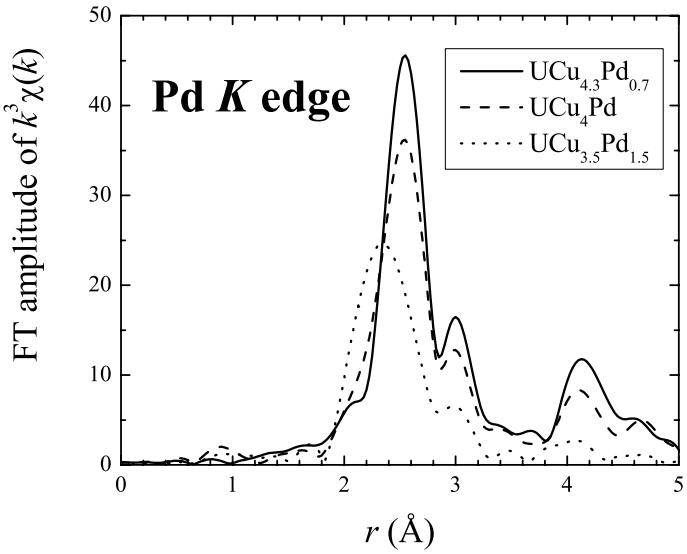

Fig. 1. Fourier transform amplitudes of $k\chi(k)$ from the Pd K edge for three of the samples used in this study. Note the clear appearance of a short peak at ≈ 2.3Å, corresponding to Pd atoms on Cu sites. The transform is from 2.5-15 Å^{-1}, and Gaussian narrowed by 0.3 Å^{-1}

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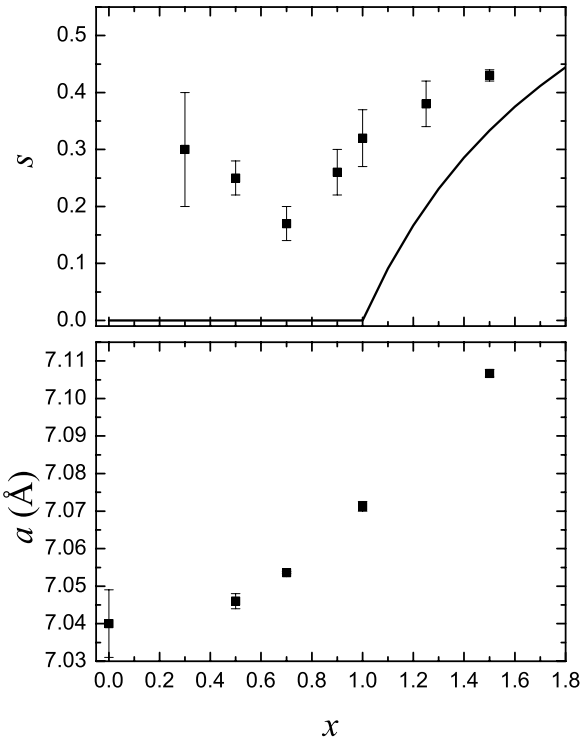

Fig. 2. The measured and nominal (line) fraction s of Pd atoms on nominally Cu ($16e$) sites is shown in the top panel, and the lattice parameters of these (or similar) samples are shown in the bottom panel for comparison.